EUROPHYSICS LETTERS 1 November 2004

Europhys. Lett., **68** (3), pp. 363–369 (2004)
DOI: 10.1209/epl/i2004-10209-3

The $^2H(d,p)^3H$ reaction in metallic media at very low energies

K. Czerski[1,2](*), A. Huke[1], P. Heide[1] and G. Ruprecht[1]

[1] *Hahn-Meitner-Institut, Bereich Strukturforschung*
Glienicker Str. 100, 14109 Berlin, Germany
[2] *Technische Universität Berlin, Institut für Atomare Physik und Fachdidaktik*
Hardenbergstr. 36, 10623 Berlin, Germany

received 5 January 2004; accepted in final form 31 August 2004
published online 16 October 2004

PACS. 25.45.Hi – Transfer reactions.
PACS. 95.30.Dr – Atomic processes and interactions.
PACS. 95.30.-k – Fundamental aspects of astrophysics.

Abstract. – Based on our experimental studies of the electron screening effect in the $^2H(d,p)^3H$ reaction for five deuteron-implanted solid targets (C, Al, Zr, Pd, Ta), theoretical calculations have been performed within an improved dielectric function theory. The theory describes correctly the observed target material dependence of the screening energies, underestimating, however, the absolute values by about a factor of 2. Applying an effective screening energy approach, the theoretical cross-sections, thick-target yields as well as nuclear reaction rates have been calculated down to the energies corresponding to the conditions of so-called cold-fusion experiments. This allows for a comparison of the experimental results at higher energies with those achieved in the heavy-water electrolysis experiments.

Introduction. – Nuclear reaction rates at very low projectile energies, far below the Coulomb barrier, are sensitive to the electronic properties of target materials. The electrons surrounding the reacting nuclei can increase the tunneling probability through the Coulomb barrier leading to an enhancement of nuclear reaction rates at low projectile energies. The electron screening effect was originally discussed due to its importance for dense astrophysical plasmas, where nuclear reaction rates can be increased by many orders of magnitudes [1]. Experimentally, the screening effect could be verified only fifteen years ago in gas target experiments [2] by an observation of an exponential-like increase of the measured cross-section for decreasing projectile energies compared to the cross-section expected for bare nuclei. Theoretically, this effect was described [3] by applying a conception of the electron screening energy resulting from the gain in the electron binding energy between the initially separated atoms and the finally united atom. In the experiments, the screening energy could be treated as an energy shift of the kinetic energy of the reacting nuclei causing an increase of the penetration probability through the Coulomb barrier.

(*) E-mail: czerski@hmi.de

TABLE I – *Electron screening energies determined for various target materials.*

Target material	Stoichiometry	Screening energy (eV) experiment	Screenining energy (eV) theory		
			Polarization	Cohesion	Total
C	CD	-20 ± 5	2.0	12.3	14.3
Al	AlD	191 ± 12	76.2	20.4	96.6
Zr	ZrD_2	295 ± 7	72.4	40.0	112.4
Pd	$PdD_{0.2}$	296 ± 15	90.6	43.2	133.8
Ta	TaD	302 ± 13	82.3	53.4	135.7

The fusion reactions $^2\mathrm{H(d,p)^3H}$ and $^2\mathrm{H(d,n)^3He}$ are especially suitable for the study of such effects, due to their relatively high cross-sections and low Coulomb barrier. Recently [4], we have shown that the screening effect investigated for the d + d fusion reactions in metallic environments is by about a factor of 10 stronger than that observed for the gas target [5] and up to a factor of 4 larger than the theoretical predictions [6]. Such findings were also confirmed by results of other groups [7–10]. From a theoretical point of view, the experiments performed with deuterized metals provide information about the electron screening not only due to bound electrons as is the case for gas-phase experiments, but also about a contribution arising from free metallic electrons. The latter gives us a possibility to test models of the electron screening based on the dielectric function theory which describes the electronic dynamics in solids [11] as well as in dense astrophysical plasmas [28]. Here, we compare our previously published [4] and some new experimental data with the results of an improved theoretical approach including the self-consistent correction of the dielectric function. This enables us to determine a reliable deuteron-deuteron potential within a host metal also for large distances corresponding to very low deuteron energies. Applying the form of the screened potential, we predict some consequences for nuclear reactions taking place in metallic environments at energies down to room temperature. By means of the theoretical calculations, we shall compare our experimental results achieved at higher energies using a classical accelerator technique with the data obtained in so-called cold-fusion experiments, *i.e.* in the heavy-water electrolysis [12, 13].

Experimental procedure. – The experimental results obtained for C, Al, Zr and Ta have already been presented previously [4, 14, 15]. The experiment with Pd has been performed analogously, so only an outline of the experimental procedure will be given here. The D^+ and D_2^+ beams from the electrostatic accelerator of energies ranging between 5 and 60 keV were magnetically analyzed, focused and impinged on a Pd foil. Differently to the previous experiment, where the targets were implanted to large deuteron densities corresponding approximately to the chemically stable stoichiometry (see table I), the implantation process of the Pd foil was interrupted at a relatively small deuteron density in order to study fusion reactions in the metallic environment possessing a small number of crystal-lattice defects and reducing the number of possible deuterium bubbles resulting from long-term irradiation. The reaction products (protons, tritons and ^{3}He particles) were detected by four Si-detectors located in the reaction plane at backward angles. Since the thickness of all targets was much larger than the projectile range, the experimental results can be expressed by the thick-target yield $Y(E)$. Here we restrict ourselves only to the reaction $^2\mathrm{H(d,p)^3H}$ for which the total thick-target yield was obtained by integrating the proton spectral line over the experimentally determined angular distribution [4]. An exponential-like enhancement of the total yields observed in the experiments for decreasing projectile energies can be described using the conception of the electron screening energy U_e [3]. According to this, a reduction of the height of the Coulomb barrier corresponds to an increase of the kinetic energy of the reacting nu-

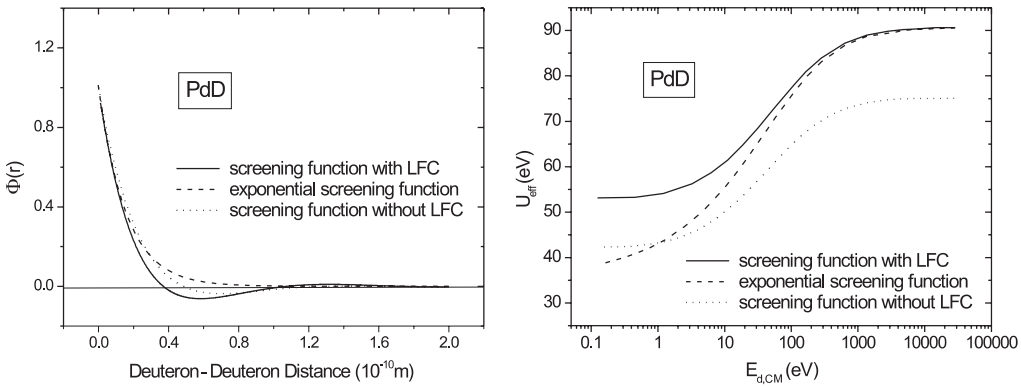

Fig. 1 – Screening function calculated with and without the local field correction (LFC) (left) and the corresponding effective screening energies (right) obtained for PdD.

clei in the expression for the barrier penetration. Consequently, a formula for the screened cross-section reads as follows:

$$\sigma_{\rm scr}(E_{\rm cm}) = \frac{1}{\sqrt{E_{\rm cm}(E_{\rm cm}+U_{\rm e})}} S(E_{\rm cm}) \exp\left[-\sqrt{\frac{E_{\rm G}}{E_{\rm cm}+U_{\rm e}}}\right]. \tag{1}$$

Here $S(E_{\rm cm})$ is the astrophysical S-factor determined for the bare nuclei taken at the center-of-mass energy $E_{\rm cm}$. $E_{\rm G} = 986\,{\rm keV}$ is the Gamow energy. In the case of $U_{\rm e} = 0$, one obtains the standard relation between the cross-section and the S-factor. Note that eq. (1) differs from the analogous formula used in the previous works [3, 4, 7–9], where the screening energy correction has been incorrectly applied also to the wavelength dependence of the cross-section being inversely proportional to the square root of E. At the very low deuteron energies investigated here this proper correction is significant.

Since the bare nuclei S-factor is very well known from the precision measurements performed on the gas target [16], the experimentally determined enhancement of thick-target yields can be described by fitting the screening energy $U_{\rm e}$. The results obtained for different targets are presented in table I. A clear target material dependence of the screening energy can be observed.

Theoretical approach —target material dependence. – The screening effect in metals can be described in analogy to the strongly coupled astrophysical plasmas. We apply here the so-called dielectric-function method [11] which allows to treat the electron screening as a static polarization of the metallic medium induced by the positively charged deuterons. The screened Coulomb potential $V(r)$ is a solution of the Poisson equation and can be expressed as a Fourier transform:

$$V(r) = \frac{e^2}{r}\Phi(r) = \frac{1}{(2\pi)^3}\int \frac{4\pi(e\varphi(q))^2}{\epsilon_\nu(q)\epsilon_c(q)q^2}\exp[i\vec{q}\vec{r}]{\rm d}^3q, \tag{2}$$

where $\epsilon_\nu(q)$ and $\epsilon_c(q)$ are the static, wave-number–dependent, dielectric functions resulting from quasi-free valence electrons and from bound metallic core electrons, respectively. Different from our previous calculations [17], the elementary charge e is multiplied by a self-consistent charge form-factor $\varphi(q)$ for deuterons with the screening electrons in the Thomas-Fermi approximation [11, 18]: $\varphi(q) = 1 - z + zq^2/(q^2 + k_{\rm TF}^2)$. Here, the Thomas-Fermi wave

number $k_{\mathrm{TF}}^2 = 6\pi e^2 n/E_{\mathrm{F}}$ has been used; n and E_{F} are the electron number density and the Fermi energy, respectively. The number z corresponds to the fraction of electrons bound to deuterons and is for metals close to unity. Since we are interested in the evaluation of the strongest possible screening effect, we uniformly set $z = 1$ for all target materials. In the absence of screening $\epsilon_\nu \equiv \epsilon_c \equiv 1$ and $z = 0$, $V(r)$ reduces to the bare Coulomb potential ($\Phi(r) \equiv 1$). The response of the valence electron gas to an external field is given by the dielectric function [19]:

$$\epsilon_\nu(q) = 1 - \frac{v(q)P(q)}{1 + v(q)G(q)P(q)}\,, \tag{3}$$

where $v(q) = 4\pi e^2/q^2$ and $P(q)$ is the static Lindhard RPA polarizability [20]. $G(q)$ is the static local field correction (LFC) that takes into account the short-range electron correlation and the exchange interaction [19]. Similar to [17], we apply for core electron polarization the dielectric function proposed in [21]. The screening function $\Phi(r)$ calculated with and without LFC by a numeric integration of eq. (2) is presented in fig. 1 together with the simple exponential function $\exp[-r/a]$ corresponding to the Bohr screening. The differences between both curves are especially visible for larger distances where the numeric potential becomes negative and shows characteristic Friedel oscillations. The attractive part of the potential is strong enough to build the hydrogen molecule. Similar results were obtained within the dielectric-function formalism [18] and the density functional theory [22]. The theoretical results were confirmed by observation of the dihydrogen complexes in many metals [23]. For distances small compared to the screening length a, the Coulomb potential is reduced by a polarization screening energy $U_{\mathrm{pol}} = e^2/a$ which is used for calculating the screened cross-section according to eq. (1).

In the metallic lattice, besides electrons also positive ions can contribute to the screening of the Coulomb barrier between reacting nuclei. This effect, called cohesion screening, can be calculated in analogy to the dense astrophysical plasmas within the ion-sphere model of Salpeter [1]. We have proposed to use a more realistic model [17] based on the universal ion-ion potential introduced by Biersack [24]. This potential describes the interaction between light as well as heavy ions at low energies with very good accuracy. Since the potential energy of two deuterons in the field of a host metal atom is larger than that of the helium atom produced in the fusion reaction, one obtains a gain in potential energy. As cohesion screening energy U_{coh} we calculate the potential energy gain resulting from the surrounding 12 host atoms. The cohesion screening energy depends on the distance between the deuteron and the host atoms and on the crystal structure. Since we are interested only in a rough estimation of this effect, we assume for all materials the same fcc crystal structure with a distance to the host atoms of 1 Å. The theoretical values for U_{e}, being a sum of the polarization and cohesion screening energies $U_{\mathrm{pol}} + U_{\mathrm{coh}}$, calculated for all target materials investigated are presented in table I.

Extrapolation towards lower energies. – For even lower projectile energies, the effect of the electron screening cannot longer be represented by an energy-independent U_{e}. We can introduce, however, an energy-dependent effective screening energy U_{eff} which ensures that the barrier penetration factors, calculated in the frame of the WKB approximation with the screened potential $V(r)$ and that applied by eq. (1), are equal to each other. This condition reads as follows:

$$\sqrt{\frac{E_{\mathrm{G}}}{E_{\mathrm{cm}} + U_{\mathrm{eff}}}}\exp\left[-\sqrt{\frac{E_{\mathrm{G}}}{E_{\mathrm{cm}} + U_{\mathrm{eff}}}}\right] = \exp\left[-\frac{2\sqrt{M}}{\hbar}\int_{R_1}^{R_2}\sqrt{V(r) - E_{\mathrm{cm}}}\,\mathrm{d}r\right], \tag{4}$$

where R_1 and R_2 are the classical turning points in the WKB expression, and M is the deuteron mass. The method proposed here allows the calculation of the cross-section according

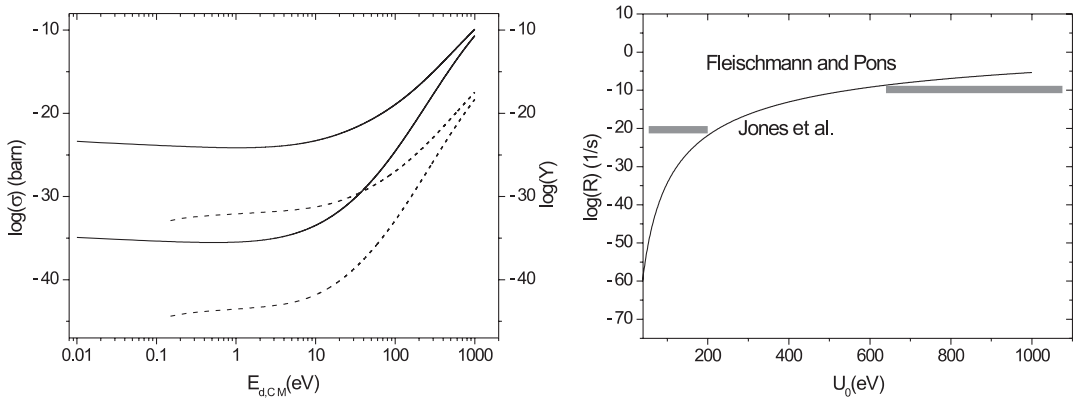

Fig. 2 – Left: energy dependence of the cross-section (full lines) and of the thick-target yield (dashed lines) for the ^{2}H(d, p)^{3}H reaction calculated for two different screening energies, 300 eV (upper lines) and 150 eV (lower lines). Right: the nuclear reaction rate in dependence on the electron screening energy. The calculations are performed for Pd.

to eq. (1) also at very low projectile energies. The effective screening energy $U_{\rm eff}$, evaluated for Pd, is shown in fig. 1. There are two well-defined limits: at high energy ($E_{\rm cm} > 1\,{\rm keV}$) and at low energy ($E_{\rm cm} < 10\,{\rm eV}$). The ratio between the low-energy and the high-energy value amounts to about 0.58, being nearly independent of the actual deuteron-deuteron potential. The total screening energy is the sum $U_{\rm eff}+U_{\rm coh}$, the value of which taken at the zero projectile energy U_0 corresponds to 0.78 of the high-energy limit $U_{\rm e}$.

Since the effective screening energy is a slowly varying function of energy, it is simple to deduce the behaviour of the cross-section at very low projectile energies. Using eq. (1), we obtain $\sigma_{\rm scr}(E_{\rm cm}) \propto 1/\sqrt{E_{\rm cm}}$ arising solely from the wavelength dependence of the cross-section. Similarly, we get an expression for the thick-target yield: $Y_{\rm scr}(E_{\rm cm}) \propto \ln(E_{\rm cm}/E_0)$, where it was taken into account that the stopping power at very low energies, being composed of the nuclear and the electronic contributions, is proportional to the square root of the energy. E_0 is the minimum energy of the deuterons in the target before they are captured in the crystal lattice, corresponding to the activation energy for deuteron diffusion in the target material. In the case of thermal equilibrium, a useful quantity for a comparison with experimental data is the nuclear reaction rate:

$$R_{\rm scr}(E_{\rm cm}) = N\sigma_{\rm scr}(E_{\rm cm})v_{\rm rel} = N\sigma_{\rm scr}(E_{\rm cm})\sqrt{\frac{4E_{\rm cm}}{M}} \cong \frac{2NS_0}{\sqrt{MU_0}}\exp\left[-\sqrt{\frac{E_{\rm G}}{U_0}}\right]. \qquad (5)$$

Here $v_{\rm rel}$ is the relative velocity between reacting deuterons, M is the deuteron mass and N denotes the deuteron density of the target. The reaction rate for projectile energies much smaller than the screening energy is dependent only on U_0 and not on $E_{\rm cm}$. Therefore, no assumption about the distribution of the deuteron velocity is necessary. The cross-section and thick-target yield for Pd involving the realistic stopping power values [25] and the effective screening energy are depicted in fig. 2. Applying eq. (5) we can also evaluate the reaction rate in dependence on the screening energy U_0. In fig. 2 a comparison with the experimental reaction rates determined in heavy-water electrolysis experiments is presented. The neutron production rates measured by Jones *et al.* [12] are by a factor 10^{10} smaller than the nuclear fusion rates determined by Fleischmann and Pons [13] from the heat excess release.

Discussion and conclusions. – The experimental electron screening energies determined for five different target materials are presented in table I together with the results of theoretical calculations. The negative U_e value for the amorphous-carbon target arises probably from the strong chemical bonding between deuterium and carbon atoms (ca. 5 eV) and a systematic uncertainty in the beam energy. The experimental screening energies for heavier metals (Zr, Pd, Ta) are of the same order, 300 eV.

The screening energy determined for Ta by the LUNA Collaboration [7] (309 ± 12 eV) and for Pd by the Japanese group [9] (310 ± 50 eV) are very close to our values, whereas the corresponding Pd value obtained by the LUNA Collaboration is much larger and amounts to 800 ± 90 eV. The lower U_e values for Al and Zr targets reported by the LUNA Collaboration [8] can be due to the enhanced oxidation of these materials under high-vacuum conditions and ion impact [4, 26]. This was also confirmed by an increase of the screening energies for Al and some other metals in the next experiment of this group [10], where the target surfaces were cleaned by Kr sputtering immediately before the deuteron incidence. However, in a vacuum of order 10^{-8} mbar the targets can re-oxidize within a few minutes (see, for example, [27]). This effect depends very strongly on the chemical reactivity of the target material and can be, on the other hand, reduced by the sputtering process of the target surface during the deuteron irradiation, which is, however, also target material dependent. Thus, the small value of $U_e = 80 \pm 20$ eV obtained for Zr and some other metals [10], being significantly smaller than both our experimental and theoretical values, might be due to the re-oxidation process of the target. Since the main goal of the present paper is a theoretical one, we refer for a detailed discussion of experimental results to our forthcoming paper [28]. Certainly, future experiments performed under ultra-high vacuum conditions will allow to clear the discrepancy.

The theoretical calculations describe the material dependence of the screening energy, observed in our experiment, qualitatively correctly. Whereas the screening energy for C is close to zero, the value for Al is slightly smaller than those calculated for the heavier metals. A strong variation of the screening energies observed in [8, 10] for metallic targets cannot be theoretically explained. The main contribution to the theoretical values is provided by polarization of the free valence electrons, although the contribution of bound electrons of the host atoms (core polarization) cannot be neglected [17]. The slight increase of U_e with the atomic number results mainly from the cohesion contribution. However, the absolute values of the theoretically calculated U_e are low by a factor of two compared with our experimental results. Including the self-consistent correction and the full wave number dependence of the dielectric function leads to lower screening energies than those determined within the simplified theory [17]. No reason for such a large discrepancy between theoretical and experimental values has been found so far. Even if a possible contribution of the channeling effect to the experimentally determined U_e were taken into account —in the case of Ta much smaller than 100 eV [15]— the difference between experiment and theory remains large. Therefore, the calculations (fig. 2) of the cross-section and the thick-target yield at extremely low energies were carried out with the experimental value of the screening energy by applying the theoretically predicted energy dependence of U_e. The resulting thick-target yield at the smallest projectile energies cannot explain the observation of the neutron emission at room temperature reported in [12, 29]. Even if we take into account that the slow projectile deuterons win kinetic energy due to adsorption at the palladium surface (ca. 4 eV [30]) and due to the electron capture in the bulk (13.6 eV), the theoretical neutron yield remains by a factor of about 10^{10} smaller than the reaction rates reported by Jones *et al.* [12]. However, the situation changes completely if we assume that the target deuterons move. Irradiation of the palladium target during the electrolysis can lead to strong lattice oscillations in the surface region of the target, causing, in turn, a quasi-free movement of target deuterons. This mechanism seems to be plausible for Pd, where the activation energy

for deuteron diffusion amounts only to 0.15 eV [30]. In this case we can use eq. (5) for the estimation of nuclear reaction rates. In fig. 2, the reaction rates calculated for different effective screening energies U_0 are compared to the neutron production rate of Jones [12] and to the heat excess rate postulated by [13]. The neutron production rate [12] requires $U_0 = 220$ eV which corresponds to a screening energy U_e of about 280 eV at the high-energy limit in agreement with our experiments. On the contrary, the energy production rate [13] demands $U_0 = 620$ eV (790 eV at high energies) being clearly outside our experimental constraints.

Summarizing, the improved dielectric function theory allows for a qualitative description of the observed target material dependence of the screening energy; the absolute values are, however, by a factor of 2 smaller than the experimental values. Applying the effective screening energy approach, cross-sections and thick-target yields could be calculated for projectile energies much lower than those achieved in our accelerator experiments. An agreement with the neutron production rates at room temperature reported by [12] could be obtained only under the assumption of quasi-free moving deuterons in the target lattice during the heavy-water electrolysis.

REFERENCES

[1] Salpeter F. E., *Austr. J. Phys.*, **7** (1954) 373.
[2] Engstler S. *et al.*, *Phys. Lett. B*, **202** (1988) 179.
[3] Assenbaum H. J., Langanke K. and Rolfs C., *Z. Phys. A*, **327** (1987) 461.
[4] Czerski K. *et al.*, *Europhys. Lett.*, **54** (2001) 449.
[5] Greife U. *et al.*, *Z. Phys. A*, **351** (1995) 107.
[6] Ichimaru S., *Rev. Mod. Phys.*, **65** (1993) 252.
[7] Raiola F. *et al.*, *Eur. Phys. J. A*, **13** (2002) 377.
[8] Raiola F. *et al.*, *Phys. Lett. B*, **547** (2002) 193.
[9] Kasagi J. *et al.*, *J. Phys. Soc. Jpn.*, **71** (2002) 2281.
[10] Bonomo C. *et al.*, *Nucl. Phys. A*, **719** (2003) 37c.
[11] Pines D., *Elementary Excitations in Solids* (Benjamin, New York) 1963.
[12] Jones S. E. *et al.*, *Nature*, **338** (1989) 737.
[13] Fleischmann M. and Pons S., *J. Electroanal. Chem.*, **261** (1989) 301.
[14] Huke A., PhD Thesis, Technische Universität Berlin (2002).
[15] Czerski K. *et al.*, *Nucl. Instrum. Methods B*, **193** (2002) 183.
[16] Brown R. E. and Jarmie N., *Phys. Rev. C*, **41** (1990) 1391.
[17] Czerski K., Huke A. and Heide P., *Nucl. Phys. A*, **719** (2003) 52c.
[18] Ichimaru S. *et al.*, *J. Phys. Soc. Jpn.*, **59** (1990) 1333.
[19] Moroni S., Ceperley D. M. and Senatore G., *Phys. Rev. Lett.*, **75** (1995) 689.
[20] Grosso G. and Parravicini G. P., *Solid State Physics* (Academic Press) 2000.
[21] Penn D. E., *Phys. Rev.*, **128** (1962) 2093.
[22] Tomanek D. *et al.*, *Phys. Rev. B*, **43** (1991) 4699.
[23] Jessopand P. G. and Morris R. J., *Coord. Chem. Rev.*, **121** (1992) 155.
[24] Ziegler J. F., Biersack J. P. and Littmark U., *The Stopping and Ranges of Ions in Solids* (Pergamon Press, New York) 1985.
[25] Ziegler J. F. and Biersack J. P., code SRIM, `http://www.srim.org`.
[26] Huke A., Czerski K. and Heide P., *Nucl. Phys. A*, **719** (2003) 279c.
[27] Czerski K. *et al.*, *Nucl. Instrum. Methods B*, **225** (2004) 72.
[28] Huke A., Czerski K. and Heide P., to be published.
[29] De Ninno A. *et al.*, *Europhys. Lett.*, **9** (1989) 221.
[30] You J. H. *et al.*, *Phys. Rev. B*, **43** (1991) 7293.